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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of: Norikazu NISHIYAMA, Yasuyuki EGASHIRA,
and Korekazu UEYAMA
Serial No.: 10/068,491 Group Art Unit: 1775
Filed: February 5, 2002 Examiner: Stein, Stephen J.
Conf. No.: 7267
For: MESOPOROUS SILICA, MESOPOROUS SILICA COMPOSITE
MATERIAL, AND PROCESSES FOR PRODUCTION THEREOF

Commission for Patents
P. O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER 37 CFR §1.132

Sir:

I, Nobuhiko Mori, a citizen of Japan, hereby declare and state:

1. I have a Doctorate degree in Engineering which was conferred upon me by Osaka University in 2000.
2. I have been employed by NGK Insulators, Ltd. since 1999. I have six years of work and research experience in the field of inorganic chemistry.
3. I am a member of The Society of Chemical Engineers, Japan
4. I am familiar with the prosecution history of the above-referenced U.S. Patent application, and have reviewed the Chaudhari prior art references cited in the Office Action mailed January 27, 2005. The mesoporous silica disclosed in Chaudhari is synthesized by forming an Si-containing gel and then adding a Zr source to the gel. Conversely, the mesoporous silica of the present invention is synthesized by mixing the Si source and the Zr source to form the gel.

5. The following experiments were conducted by me or under my direct supervision to show that mesoporous silica formed by mixing the Si source and Zr source to form a gel has a patentably distinct structure (that is exemplified at least by an improved alkaline resistance) compared to mesoporous silica having an identical composition but which is instead formed by adding the Zr source after the Si-containing gel is initially formed.

The following reagents were used in both Experiment A (Inventive Example) and Experiment B (Comparative Example):

C ₁₆ TAB	(cetyltrimethyl ammonium bromide);
TEOS	(tetraethylorthosilicate); and
ZrPr	(Zirconium propoxide).

The above reagents were used in the following molar ratios in both Experiment A and Experiment B:

C ₁₆ TAB	0.6;
TEOS	1.0;
ZrPr	0.03;
NaOH	0.5; and
H ₂ O	61.

EXPERIMENT A (Inventive Example):

The mesoporous silica samples of Experiment A (the Experiment A samples) were produced as follows:

A mixed solution containing both TEOS (i.e., the Si source) and ZrPr (i.e., the Zr source) was prepared, and then the solution was mixed for 2 hours at 303° K with another solution containing C₁₆TAB (i.e., a surfactant) to form a gel. The gel was then processed to form a mesoporous silica by placing in a pressure vessel kept at a predetermined temperature.

Five samples were produced, and the structure of the Experiment A samples were

evaluated by x-ray diffraction (using the 211 and 220 phases as standard axes)

before the Experiment A samples were subjected to alkaline resistance testing in the manner described below.

The alkaline resistance of the mesoporous silica samples produced according to Experiment A was tested and evaluated in the following manner:

A plurality of alkaline test solutions, each having a predetermined pH value in a range of 10 to 11.5, were prepared by dissolving NaHCO_3 and NaOH in appropriate amounts. Each of the Experiment A samples was immersed in a respective one of the alkaline test solutions for 3 hours at a temperature of 30EC. The samples were dried at 363° K for 12 h, and then the structure of each sample was again evaluated using the above described x-ray diffraction technique to determine the alkaline resistance of the Experiment A samples.

EXPERIMENT B (Comparative Example):

The mesoporous silica samples of Experiment B (the Experiment B samples) were produced as follows:

A solution containing TEOS (i.e., the Si source) and (i.e., a surfactant) was mixed for 2 h at 303° K (temperature) to form an initial gel. The initial gel was then mixed with ZrPr (i.e., the Zr source) for 2 h (time) 303° K to form a final gel. The final gel was then processed to form a number of mesoporous silica samples in the same manner as described above in connection with Experiment A.

The alkaline resistance of the Experiment B samples was tested and evaluated in the same manner described above in connection with Experiment A samples.

RESULTS

Peak intensities of [211] and [220] of the Experiment A samples showed no significant changes after the alkali treatment in the alkali solutions having a pH of 10 to 11.5. On the other hand, peak intensities of [211] and [220] of the Experiment B samples significantly decreased, that is, substantially disappeared after the alkali treatment in the alkaline solution of pH 10. These results show that the Experiment A samples had higher alkali resistance compared to the Experiment B samples.

6. The above results show that, when a Zr source is provided along with an Si source in a solution or dispersion that is then mixed with a surfactant to form a gel, the evaluated alkaline resistance of the resultant mesoporous silica was high. To the contrary, when the Zr source is provided after an initial gel is formed from a solution based on an Si source and a surfactant, the evaluated alkaline resistance of the resultant mesoporous silica was low. In that manner, it is clear to me that the sequence of the Zr source introduction, that is, the point in time at which the Zr source is introduced during the formation process (i.e., with the Si source before the gel is initially formed versus after the gel is initially formed by mixing the Si source with a surfactant) affects the resultant structure of the mesoporous silica. This is evidenced, for example, by the clear differences in the evaluated alkaline resistance properties of the experimental samples.

7. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

April 22, 2005
Date

Nobuhiko Mori
Nobuhiko MORI